Ultramicroscopy ■ (■■■■) ■■■–■■■

Contents lists available at ScienceDirect

Ultramicroscopy

journal homepage: www.elsevier.com/locate/ultramic

Analysis of grain boundary dynamics using event detection and cumulative averaging

A. Gautam [a], C. Ophus [a], F. Lançon [b], P. Denes [a], U. Dahmen [a,*]

[a] National Center for Electron Microscopy, LBNL, Berkeley, CA 94720, USA
[b] Laboratoire de Simulation Atomistique (L_Sim), SP2M, INAC, CEA, 38054 Grenoble, France

ARTICLE INFO

Article history:
Received 27 August 2014
Received in revised form
6 November 2014
Accepted 6 November 2014

Keywords:
HRTEM
HAADF
Dynamics
Atomic structure
Grain boundary
Simulation
Step
Time average
Aberration correction
Electron detector

ABSTRACT

To analyze extended time series of high resolution images, we have employed automated frame-by-frame comparisons that are able to detect dynamic changes in the structure of a grain boundary in Au. Using cumulative averaging of images between events allowed high resolution measurements of the atomic relaxation in the interface with sufficient accuracy for comparison with atomistic models. Cumulative averaging was also used to observe the structural rearrangement of atomic columns at a moving step in the grain boundary. The technique of analyzing changing features in high resolution images by averaging between incidents can be used to deconvolute stochastic events that occur at random intervals and on time scales well beyond that accessible to single-shot imaging.

1. Introduction

The motion of interfaces in solids is often controlled by the nucleation and propagation of steps [1–3]. However, nucleation events are stochastic in nature and thus difficult to follow dynamically by in-situ observation. For example the migration of grain boundaries during capillary shrinkage of island grains in Au typically follows non-parabolic kinetics and proceeds erratically, so that dormant periods alternate with rapid bursts of activity or structural changes [4].

Observations of interface dynamics are usually made by diffraction contrast or high resolution imaging at video rates [5–9]. This limits the spatial and temporal resolution of the observations to features that are large enough to be resolved and slow enough to be recorded. These constraints are particularly restrictive for stochastic events such as step motion or fluctuation between different metastable states. By their nature, such events occur at random intervals and thus do not lend themselves to time-resolved pump-probe experiments or stroboscopic techniques [10,11]. The fast rate of such transitions usually makes it impossible to record them directly, and thus high resolution images often represent a superposition of different structural states, resulting in image blurring of the critical features, even in atomic resolution images with high S/N ratio.

However, thanks to recent advances in electron microscopy, it has become possible to record high resolution images with improved S/N ratio and at higher rates than previously feasible. Here we show how it is possible to deconvolute stochastic events by applying statistical averaging techniques to time-resolved image sequences. This allows us to analyze the evolution of atomic structure with a precision that is sufficient for comparison with atomistic simulations.

Statistical techniques of image processing are commonly used to enhance the S/N in high-resolution imaging, typically by averaging over periodically spaced features in the spatial domain, such as structural units in a grain boundary (e.g. [12]), unit cells in a beam sensitive crystal (e.g. [13]), or randomly-distributed features such as proteins for single-particle analysis in cryo-electron microscopy [14]. In the temporal domain, statistical averaging is most commonly used in pump-probe experiments, which allow cumulative data acquisition from many low-dose images or diffraction patterns [11,15].

In the present study, we investigate the atomic structure of the (001)/(110) grain boundary in 90° ⟨110⟩ bicrystals of Au. This boundary is incommensurate because the repeat periods of lattice planes of the two grains that are aligned across the boundary are

* Corresponding author.
E-mail address: udahmen@lbl.gov (U. Dahmen).

http://dx.doi.org/10.1016/j.ultramic.2014.11.008
0304-3991/

in the ratio of $1:\sqrt{2}$. For gold bicrystals, an analysis based on the Frenkel–Kontorova model found atomic relaxations characteristic of interfaces capable of superglide behavior [16–18].

Because of a strong anisotropy in the energy of this grain boundary, its migration is controlled by the nucleation and propagation of steps [3]. In order to understand this boundary in more detail, we have analyzed its dynamic behavior by following the motion of defects during observation by atomic resolution electron microscopy at room temperature. Thus, even though 300 kV electrons are well below the knock-on threshold for Au [19], the dynamic effects observed here are stimulated by the electron beam [6]. As both grains in the bicrystal have a ⟨110⟩ surface, surface energy does not provide a driving force for boundary motion, and as a result, the observed features are most likely fluctuations around equilibrium. However, such structural fluctuations are expected to be important for thermally activated processes during grain coarsening, step motion and deformation.

2. Experimental procedure

The samples were prepared by physical vapor deposition of high purity Au onto heated single crystal Ge substrates [20]. Due to preferred heteroepitaxial alignment, thin films grew as two symmetry-equivalent ⟨110⟩ variants, rotated 90° relative to each other in the mazed bicrystal structure, with grain boundaries of fixed misorientation but variable inclination. The Ge substrate was later dissolved in a 1:1 solution of hydrofluoric acid and hydrogen peroxide, resulting in freestanding Au films of about 4 nm thickness.

Thin film samples were imaged by high resolution electron microscopy in the TEAM microscope [15], using a direct electron detector [21,22], and analyzed with custom routines written in the MATLAB programming language. Dynamic structural changes were detected by applying an edge detection filter in the time domain, using the procedure described in the Appendix.

High resolution images were recorded in the aberration-corrected TEAM microscope at 300 kV, using both, HRTEM and HAADF imaging. HRTEM images were recorded with the TEAM detector, a direct electron detector developed to optimize the efficiency and dynamic range of electron detection in aberration-corrected electron microscopes [21,22]. The detector was used at 90 frames/s and 1024×512 pixels per frame. The microscope was operated at 300 kV with $Cs = -10$ μm and $Cc < 20$ μm (typically ~5 μm). In HAADF, rapid scans were obtained at a resolution of 32 pixels/nm and a dwell time of 0.8 μs/pixel.

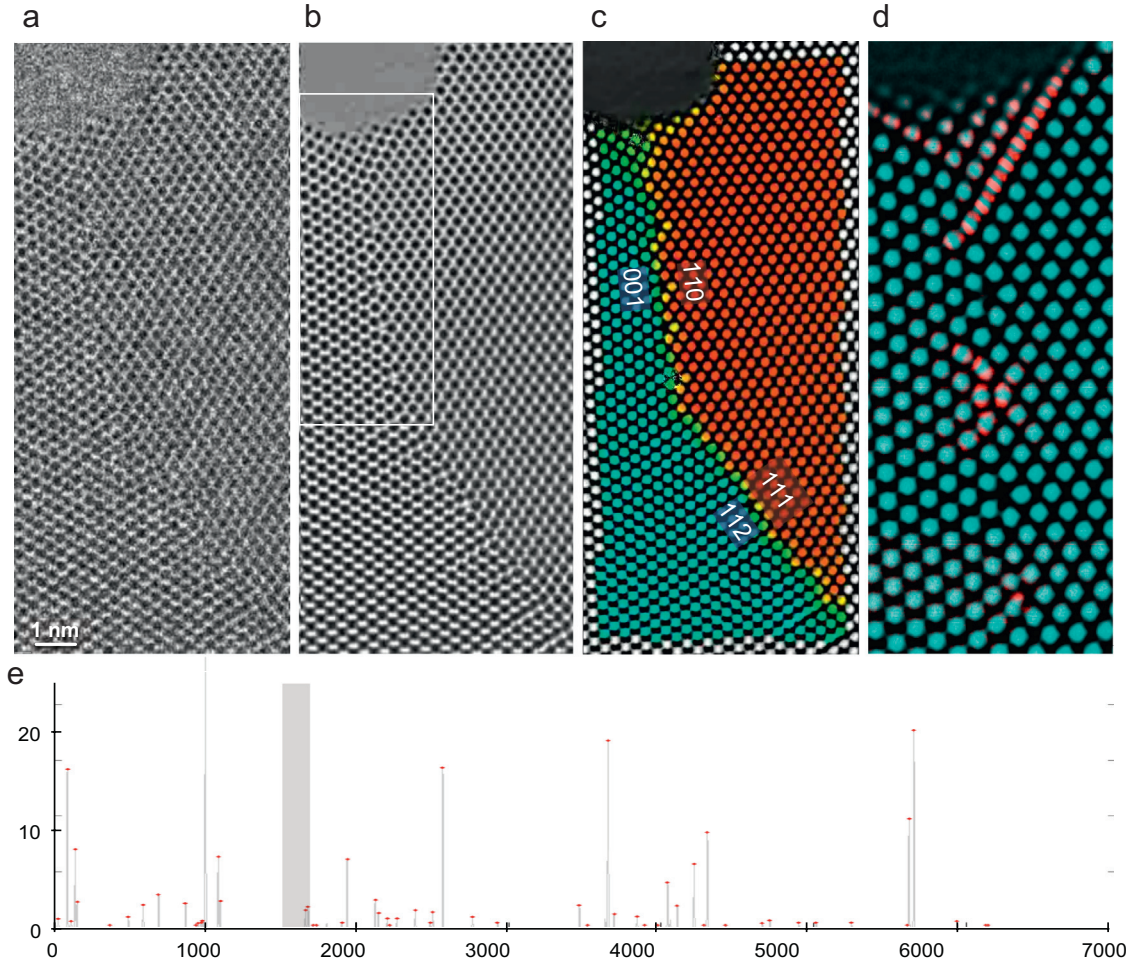

Fig. 1. Application of event detection and cumulative averaging to an HRTEM image series of 6000 frames recorded with a direct electron detector at a rate of 90 fps. An individual 512×1024 image from this series is shown in (a). Cumulative averaging between events yields high quality images (b), which can be used to analyze the nature of atomic motion by highlighting changes in local coordination (c) and local motion of atomic columns (d) in a magnified view of the (001)/(110) boundary segment framed in (b). Automated comparison between sequential images detects structural events as spikes in the correlation coefficient plotted over the entire sequence (e). The gray bar illustrates that a 2 s exposure would tend to straddle one or several events, thus convoluting different states in a single image.

Please cite this article as: A. Gautam, et al., Analysis of grain boundary dynamics using event detection and cumulative averaging, Ultramicroscopy (2014), http://dx.doi.org/10.1016/j.ultramic.2014.11.008